

# Incommensurate Phase of a Triangular Frustrated Heisenberg Model Studied via Schwinger-Boson Mean-Field Theory

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**Abstract.** We study a triangular frustrated antiferromagnetic Heisenberg model with nearest-neighbor interaction  $J_1$  and third-nearest-neighbor interactions  $J_3$  by means of Schwinger-boson mean-field theory. It is shown that an incommensurate phase exists in a finite region in the parameter space for an antiferromagnetic  $J_3$  while  $J_1$  can be either positive or negative. A detailed solution is presented to disclose the main features of this incommensurate phase. A gapless dispersion of quasiparticles leads to the intrinsic  $T^2$ -law of specific heat. The local magnetization is significantly reduced by quantum fluctuations (for  $S = 1$  case, a local magnetization is estimated as  $m = \langle S_i \rangle \approx 0.6223$ ). The magnetic susceptibility is linear in temperature at low temperatures. We address possible relevance of these results to the low-temperature properties of  $\text{NiGa}_2\text{S}_4$ . From a careful analysis of the incommensurate spin wave vector, the interaction parameters for  $\text{NiGa}_2\text{S}_4$  are estimated as,  $J_1 \approx -3.8755\text{K}$  and  $J_3 \approx 14.0628\text{K}$ , in order to account for the experimental data.

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## 1. Introduction

In two-dimensional (2D) antiferromagnets, it was proposed the "geometrical frustration" may enhance the quantum spin fluctuation and suppress the magnetic order to form a spin liquid [1]. In this context the triangular- and kagomé-related lattices are studied extensively to seek quantum spin liquid [2]. It turns out that the triangular lattice antiferromagnet with nearest-neighbor (NN) coupling exhibits  $120^\circ$  magnetic order [3], while the kagomé lattice antiferromagnet is still a controversial topic for intriguing exploring [4]. People resort to other interactions, such as longer range and multiple-spin exchange ones, to realize quantum spin liquid [2]. Experimental evidences in favor of this long-predicted spin-liquid state have emerged in recent years [5], although many aspects are still elusive. The spin disorder at low temperatures found in the compound  $\text{NiGa}_2\text{S}_4$ , in which Ni spins ( $S = 1$ ) forms a stack of triangular lattices, aroused much attention [6, 7, 8, 9]. The crystal structure of the material is highly 2D, since inter-layer interactions are quite weak. Intriguing low-temperature properties of this material include  $T^2$ -law of specific heat, incommensurate short-range spin correlation, and lack of divergent behavior of the magnetic susceptibility. A dominant third-nearest-neighbor (3rd-NN) antiferromagnetic (AFM) interaction  $J_3$  could produce the incommensurate phase in a rough picture: four sublattices will form commensurate  $120^\circ$  magnetic order separately if the NN interaction  $J_1$  is zero, and the system will be driven into an incommensurate order if  $J_1$  is gradually switched on. A first-principle calculation by Mazin [10] suggests a large 3rd-NN interaction  $J_3$  and a negligible 2nd-NN interaction.  $J_3$  is confirmed to be AFM, but the sign of  $J_1$  has not yet been identified [10]. The classical spin version of this model was studied in a Monte-Carlo simulation [11], which provides some helpful informations such as the incommensurability. Up to now, the quantum spin version of this model has not yet been studied very well. Besides the sign of  $J_1$ , many aspects of this model, either in agreement or disagreement with the experiment of  $\text{NiGa}_2\text{S}_4$ , need further clarification and treatments. In this paper we focus on the low-temperature properties of the quantum spin model and intend to make a contribution to this topic.

The Schwinger-boson mean-field theory (SBMFT) provides a reliable description for both quantum ordered and disordered antiferromagnets based on the picture of the resonant valence-bond (RVB) state [1, 12, 13]. As a merit, it does not prescribe any prior order for the ground state in advance, which should emerge naturally if the Schwinger bosons condense in the lowest energy states. For the Heisenberg antiferromagnets with NN couplings at zero temperature, it successfully captures the  $(\pi, \pi)$  magnetic order on the square lattice and the  $120^\circ$  magnetic order on triangular lattice respectively [12, 13, 14, 15]. By means of SBMFT, we will show that the  $J_1$ - $J_3$  model falls into an incommensurate order phase at zero temperature for an AFM  $J_3$  and either a FM  $J_1$  or an AFM  $J_1$ . By analyzing the incommensurate spin wave vector, we find that the NN interaction  $J_1$  should in the FM region to obtain an appropriate incommensurate phase. We also show that the  $T^2$ -law of specific heat is an intrinsic feature of this

phase, the magnetic susceptibility is linear in temperature, and the local magnetization is significantly reduced by quantum fluctuations. We address possible relevance of these results to low temperature properties of  $\text{NiGa}_2\text{S}_4$ . Our results suggests that the  $J_1$ - $J_3$  model is an essential part of the minimal model for  $\text{NiGa}_2\text{S}_4$ . In the following, we first present a formalism of the SBMFT scheme for the  $J_1$ - $J_3$  model, then solve the mean-field equations numerically and calculate relevant quantities. Finally we discuss the physical meanings of the results.

## 2. The Schwinger-boson mean-field theory

The  $J_1$ - $J_3$  model on the triangular lattice reads

$$H = J_1 \sum_{\langle ij \rangle \in \text{NN}} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle i'j' \rangle \in \text{3rd-NN}} \mathbf{S}_{i'} \cdot \mathbf{S}_{j'}. \quad (1)$$

We set  $J_3 > 0$ , but  $J_1$  can be either AFM or FM. In the Schwinger-boson representation for the spin operators,  $S_i^+ = a_i^\dagger b_i$ ,  $S_i^- = b_i^\dagger a_i$ ,  $S_i^z = (a_i^\dagger a_i - b_i^\dagger b_i)/2$  with  $[a_i, a_j^\dagger] = [b_i, b_j^\dagger] = \delta_{ij}$ , we decompose the NN and 3rd-NN interactions as[16]

$$J_1 \mathbf{S}_i \cdot \mathbf{S}_j = J_1 : F_{ij}^\dagger F_{ij} : - J_1 A_{ij}^\dagger A_{ij}, \quad (2)$$

$$J_3 \mathbf{S}_{i'} \cdot \mathbf{S}_{j'} = -J_3 \Pi_{i'j'}^\dagger \Pi_{i'j'}, \quad (3)$$

with  $F_{ij} = (a_i^\dagger a_j + b_i^\dagger b_j)/2$ ,  $A_{ij} = (a_i b_j - b_i a_j)/2$ , and  $\Pi_{i'j'} = (a_{i'} b_{j'} - b_{i'} a_{j'})/2$ . Correspondingly, we introduce three competing mean fields,  $F = \langle F_{ij} \rangle$ ,  $A = -i \langle A_{ij} \rangle$ , and  $\Pi = -i \langle \Pi_{i'j'} \rangle$ , and apply the Hartree-Fock decompositions for the interactions. A Lagrangian multiplier  $\lambda$  is also introduced to impose the constraint on the Schwinger bosons,  $+\lambda \sum_i (a_i^\dagger a_i + b_i^\dagger b_i - 2S)$ . After performing the Fourier's transform, the effective Hamiltonian can be written in a compact form,

$$H_{eff} = \sum_{\mathbf{k}} \phi_{\mathbf{k}}^\dagger M(\mathbf{k}) \phi_{\mathbf{k}} + \varepsilon_0, \quad (4)$$

where  $\phi_{\mathbf{k}}^\dagger = (a_{\mathbf{k}}^\dagger, b_{\mathbf{k}}^\dagger, a_{-\mathbf{k}}, b_{-\mathbf{k}})$ ,  $M(\mathbf{k}) = \epsilon(\mathbf{k}) \sigma_0 \otimes \sigma_0 + \Delta(\mathbf{k}) \sigma_y \otimes \sigma_y$ ,  $\epsilon(\mathbf{k}) = \lambda - J_1 F \sum_{\delta} \cos k^{(\delta)}$ ,  $\Delta(\mathbf{k}) = J_1 A \sum_{\delta} \sin k^{(\delta)} + J_3 \Pi \sum_{\delta} \sin 2k^{(\delta)}$ ,  $\varepsilon_0 = 3N_{\Lambda}(-J_1 F^2 + J_1 A^2 + J_3 \Pi^2) - N_{\Lambda} \lambda (2S + 1)$ , and  $\otimes$  means the Kronecker product,  $\sigma_0$  is a  $2 \times 2$  unit matrix,  $\sigma_{\alpha}$ 's ( $\alpha = x, y, z$ ) are Pauli matrices,  $k^{(\delta)} = k_x, k_x/2 + \sqrt{3}k_y/2, -k_x/2 + \sqrt{3}k_y/2$  for  $\delta = 1, 2, 3$  respectively. The Matsubara Green's function are defined as,

$$G(\mathbf{k}, \tau) = - \left\langle T_{\tau} \phi_{\mathbf{k}}(\tau) \phi_{\mathbf{k}}^\dagger \right\rangle, \quad (5)$$

where  $\tau$  is the imaginary time and  $\phi_{\mathbf{k}}(\tau) = e^{\tau H_{eff}} \phi_{\mathbf{k}} e^{-\tau H_{eff}}$ . All physical quantities can be expressed in terms of the matrix elements of the Green's function.

The Matsubara Green's function in Matsubara frequency  $\omega_n = 2n\pi/\beta$  ( $n$  is an integer for bosons) can be worked out as

$$G(\mathbf{k}, i\omega_n) = \frac{i\omega_n \sigma_z \otimes \sigma_0 - \epsilon(\mathbf{k}) \sigma_0 \otimes \sigma_0 + \Delta(\mathbf{k}) \sigma_y \otimes \sigma_y}{(i\omega_n)^2 - \omega^2(\mathbf{k})}. \quad (6)$$

From the poles of the Matsubara Green's function, the two degenerate spectra of the quasi-particles can be readily read out,

$$\omega(\mathbf{k}) = \sqrt{\epsilon^2(\mathbf{k}) - \Delta^2(\mathbf{k})}. \quad (7)$$

The mean-field equations are established by the constraint and the introduced mean fields. We omit the details and only present the results here,

$$\frac{1}{N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_B[\omega(\mathbf{k})]) \frac{\epsilon(\mathbf{k})}{\omega(\mathbf{k})} = 2S + 1, \quad (8a)$$

$$\frac{1}{6N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_B[\omega(\mathbf{k})]) \frac{\epsilon(\mathbf{k}) \sum_{\delta} \cos k^{(\delta)}}{\omega(\mathbf{k})} = F, \quad (8b)$$

$$\frac{1}{6N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_B[\omega(\mathbf{k})]) \frac{\Delta(\mathbf{k}) \sum_{\delta} \sin k^{(\delta)}}{\omega(\mathbf{k})} = A, \quad (8c)$$

$$\frac{1}{6N_\Lambda} \sum_{\mathbf{k}} (1 + 2n_B[\omega(\mathbf{k})]) \frac{\Delta(\mathbf{k}) \sum_{\delta} \sin 2k^{(\delta)}}{\omega(\mathbf{k})} = \Pi, \quad (8d)$$

where  $n_B[\omega(\mathbf{k})] = [e^{\omega(\mathbf{k})/k_B T} - 1]^{-1}$  is the Bose-Einstein distribution function. In the thermodynamical limit  $N_\Lambda \rightarrow \infty$ , the momentum sum is replaced by an integral,  $(1/N_\Lambda) \sum_{\mathbf{k}} \rightarrow (1/A_{BZ}) \int d^2k$ ,  $A_{BZ} = 8\pi^2/\sqrt{3}$ . If the Schwinger bosons condensation occurs at  $\mathbf{k}^*$ , a condensation term should be extracted in the momentum summation of the first equation, Eq. (8a),

$$2S + 1 = \rho_0 + \int \frac{d^2k}{A_{BZ}} (1 + 2n_B[\omega(\mathbf{k})]) \frac{\epsilon(\mathbf{k})}{\omega(\mathbf{k})}, \quad (9)$$

where the density of condensates

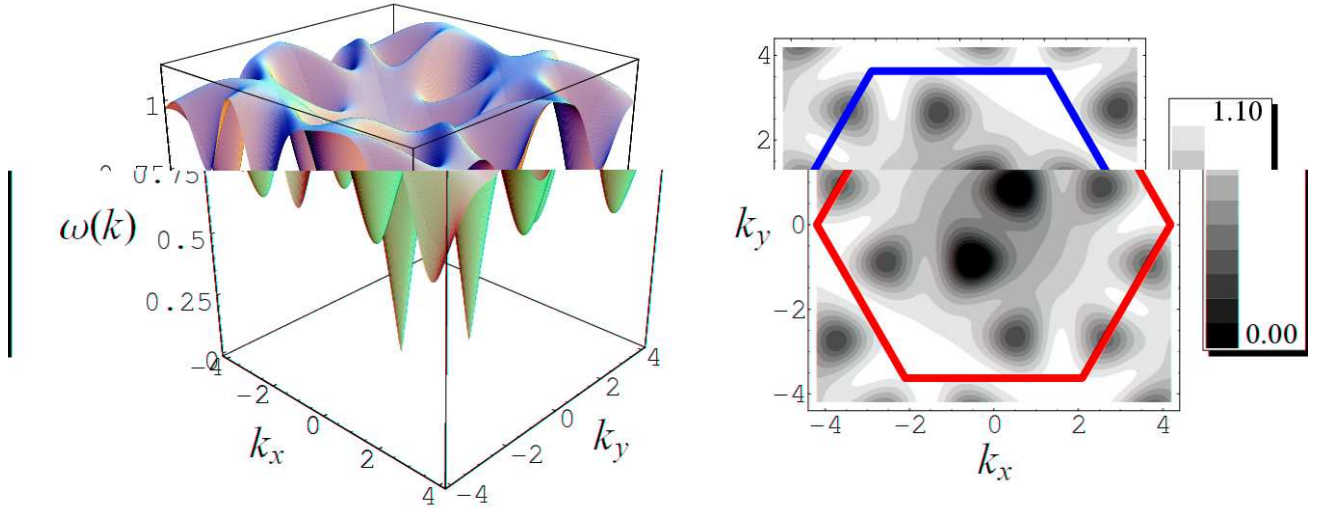
$$\rho_0 = \frac{1}{N_\Lambda} \sum_{\mathbf{k}^*} (1 + 2n_B[\omega(\mathbf{k}^*)]) \frac{\epsilon(\mathbf{k}^*)}{\omega(\mathbf{k}^*)}. \quad (10)$$

Our numerical solution demonstrates the condensation occurs at zero temperature for spin  $S > S_C$  with  $S_C \lesssim 0.172$ . Thus we will count condensations in later discussions of this paper. The condensation terms in the next three mean-field equations, Eq. (8b)-(8d), should also be extracted carefully. It is noticeable the per site ground state energy can be simplified by utilizing the mean-field equations,

$$E_0/N_\Lambda = \frac{1}{N_\Lambda} \left( \sum_{\mathbf{k}} \omega(\mathbf{k}) + \varepsilon_0 \right) = -3J_1(A^2 - F^2) - 3J_3\Pi^2 \quad (11)$$

### 3. The incommensurate phase solution

The mean-field equations are solved numerically at zero temperature. For our purpose, we set  $S = 1$  in the calculation in order to compare the result with the related experiment, although the qualitative conclusion is spin-independent, but the quantitative results vary with the values of spin. One fact that should be noticed is that the mean fields  $F$  and  $A$  could not exist simultaneously [14, 17], so the number of



**Figure 1.** (Color online) The gapless spectrum with nodal points. To compare with the experiment, we choose parameter  $J_1/J_3 = -0.2756$ , so that the gapless nodal points occur at  $\mathbf{k}^* = \pm(k^*/2, \sqrt{3}k^*/2)$  with  $k^* = 0.158\pi$ . The blue hexagon denotes the first Brillouin zone. See more details in the text.

mean-field equations can be reduced from 4 to 3 in both  $J_1 > 0$  and  $J_1 < 0$  regions. In the two regions, we found the system falls into the incommensurate phases with gapless excitations.

The quasiparticle's spectra become gapless at the nodal points, say  $\mathbf{k}^* = (k_x^*, k_y^*) = \pm(k^*/2, \sqrt{3}k^*/2)$  (e.g. see Fig. 1). Near the nodal points, the spectrum is linear in  $|\mathbf{k} - \mathbf{k}^*|$ ,

$$\omega(\mathbf{k}) \approx \alpha |\mathbf{k} - \mathbf{k}^*| + O(|\mathbf{k} - \mathbf{k}^*|^2). \quad (12)$$

At a finite temperature, a gapful spectrum will develop asymptotically as  $\Delta_{\text{gap}} = c_1 e^{-c_2/T}$  with constants  $c_1$  and  $c_2$ , which coincides with the Mermin-Wagner theorem [13]. The incommensurate order at zero temperature of the system is signalled by the divergence in the static spin structure factor,

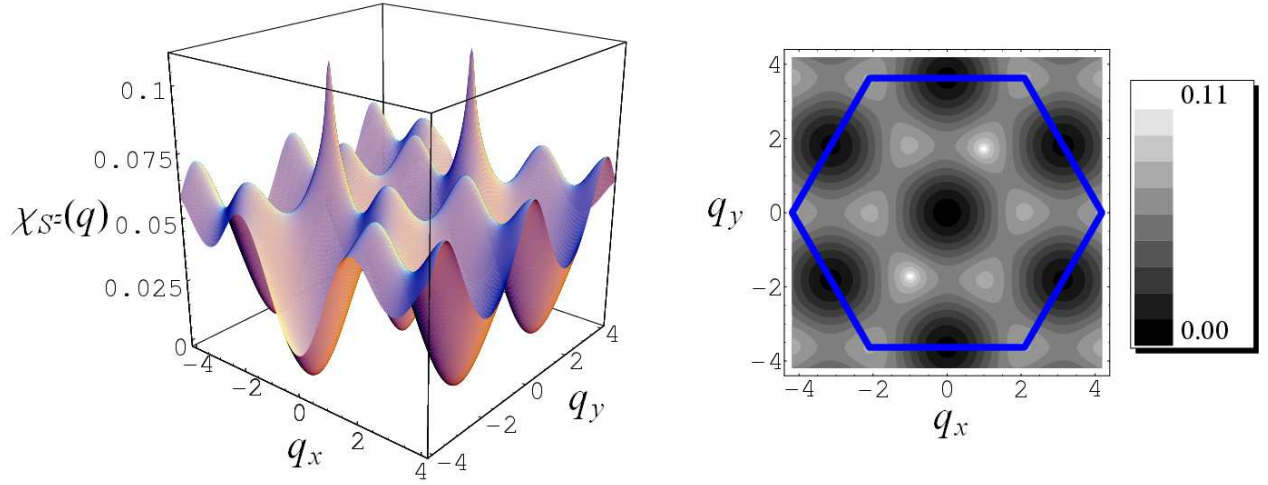
$$\chi_{S^z}(\mathbf{q}) = \frac{1}{N_\Lambda} \sum_{\mathbf{k}} \frac{1}{2} [P(\mathbf{k} + \mathbf{q}) Q(\mathbf{k}) - R(\mathbf{k} + \mathbf{q}) R(\mathbf{k})], \quad (13)$$

where  $P(\mathbf{k}) = [\epsilon(\mathbf{k})/\omega(\mathbf{k}) + 1]/2$ ,  $Q(\mathbf{k}) = [\epsilon(\mathbf{k})/\omega(\mathbf{k}) - 1]/2$ ,  $R(\mathbf{k}) = \Delta(\mathbf{k})/[2\omega(\mathbf{k})]$ . Because the spectra is gapless at  $\mathbf{k}^*$ ,  $\omega(\mathbf{k}^*) = 0$ ,  $\chi_{S^z}(\mathbf{q})$  becomes divergent at  $\mathbf{q}^* = 2\mathbf{k}^*$  (see Fig. 2),

$$\chi_{S^z}(\mathbf{q}^*) = \frac{1}{16} N_\Lambda \rho_0^2, \quad (14)$$

as it is proportional to the number of lattice sites  $N_\Lambda$ . The local magnetization will be reduced significantly due to strong quantum fluctuations,

$$m \approx \sqrt{\frac{\chi_{S^z}(\mathbf{q}^*)}{N_\Lambda |\cos \mathbf{q}^*|}} = \frac{\rho_0}{4\sqrt{|\cos \mathbf{q}^*|}}. \quad (15)$$



**Figure 2.** (Color online) The zero-temperature static spin structure factor at the parameter  $J_1/J_3 = -0.2756$ . The blue hexagon denotes the first Brillouin zone. The divergent peaks located at  $\mathbf{q}^* = 2\mathbf{k}^*$  indicate an incommensurate order.

The important difference between the regions of  $J_1 > 0$  and  $J_1 < 0$  is the nodal point's momentum  $k^* \in [\pi/6, \pi/3]$  for  $J_1 > 0$  and  $k^* \in [0, \pi/6]$  for  $J_1 < 0$  regions, respectively. In the limit of  $J_1/J_3 \rightarrow \infty$ ,  $k^* \rightarrow \pi/3$ , the solution reproduces  $120^\circ$  spin order correctly. While below the critical value  $J_1/J_3 \approx -3.71$ , the system becomes a saturated ferromagnet, where the linear expansion, Eq. (12), will be replaced by a parabolic form  $\omega(\mathbf{k}) \approx \beta(\mathbf{k} - \mathbf{k}^*)^2$ . The plots of  $k^*$  versus  $J_1/J_3$  and  $\alpha$  versus  $J_1/J_3$  are shown in Fig. 3.

The incommensurate spin wave vector observed in  $\text{NiGa}_2\text{S}_4$  is  $k^* \cong 0.158\pi < \pi/6$ . From this data we estimate that  $J_1/J_3 \approx -0.2756$  from Fig. 3, which is slight different from the value  $-0.20$  in Ref.[6], *i.e.* we have a considerable FM  $J_1$ . Thus we can exclude the possibility of AFM  $J_1$  [10]. The local magnetization at this point evaluated by Eq. (15) is  $0.6223$ , (not  $S = 1$ ), while the experimental data of  $\text{NiGa}_2\text{S}_4$  suggest a larger value,  $0.75(8)$  [6].

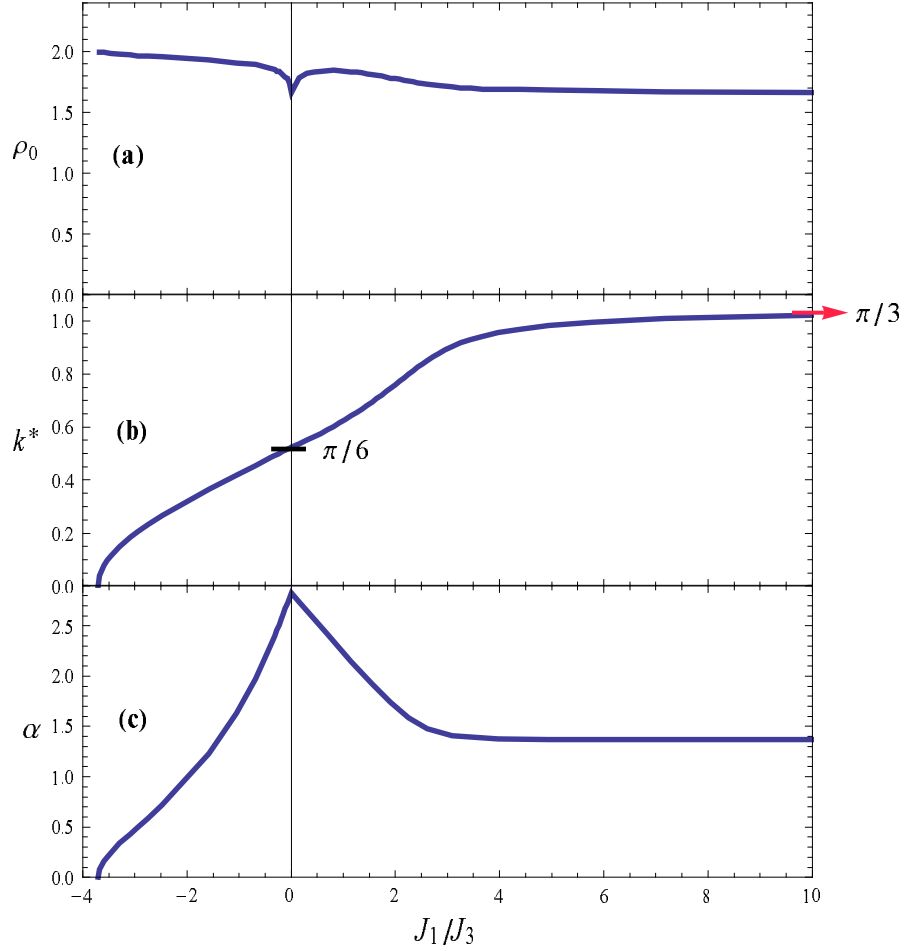
The nodal structure of the spectra, Eq. (12), leads to a linear density-of-states (DOS) in energy  $E$ ,

$$D(E) = 2 \sum_{\mathbf{k}} \delta(E - \omega(\mathbf{k})) \approx \frac{\sqrt{3}}{\pi\alpha^2} E. \quad (16)$$

where the factor 2 comes from the degeneracy of the quasiparticle spectra. As a result, a  $T^2$ -law of specific heat follows apparently,

$$C_V/N_\Lambda \approx \frac{6\sqrt{3}\zeta(3)k_B^3}{\pi\alpha^2 J_3^2} T^2, \quad (17)$$

where  $\zeta(3) = 1.202$ . If one supposes that the  $T^2$ -law of specific heat of  $\text{NiGa}_2\text{S}_4$  is ascribed to the gapless incommensurate phase, a numerical estimation,  $J_1 \approx -3.8755\text{K}$  and  $J_3 \approx 14.0628\text{K}$ , could be obtained. This result is reasonable compared to the experimental estimation  $J_3 \cong 30\text{K}$  [18].



**Figure 3.** (Color online) (a) The condensation term  $\rho_0$  versus  $J_1/J_3$ . (b) The magnitude of the nodal point's momentum of the spectrum  $k^*$  versus  $J_1/J_3$ . In the limit  $J_1/J_3 \rightarrow +\infty$ , the result reproduces the  $120^\circ$  commensurate spin order correctly. The incommensurate spin wave vector observed in  $\text{NiGa}_2\text{S}_4$ ,  $k^* \cong 0.158\pi$ , lies in the  $J_1 < 0$  region. Please see more details in the text. (c) The coefficient  $\alpha$  in Eq. (12) versus  $J_1/J_3$ .

#### 4. Discussions

Before ending this paper, we point out that the zero-field susceptibility for this incommensurate phase is linear in temperature,

$$\chi_M/N_\Lambda \approx \frac{\sqrt{3}(g\mu_B)^2 k_B T}{2\pi\alpha^2 J_3^2}. \quad (18)$$

Using the parameters noted above, we find that it is  $\chi_M \approx 2.77 \times 10^{-4} T$  (emu/mole), which is not in agreement with the experimental data of  $\text{NiGa}_2\text{S}_4$ ,  $\chi_M \approx A + BT$  with  $A \approx 0.009$  (emu/mole) and  $B \approx 0$  below 10K [6]. The Monte-Carlo study also shows the classical version of this model only produce a single peak in the specific heat [11]. These facts indicate that the model in Eq. (1) may not account for all mysteries in  $\text{NiGa}_2\text{S}_4$ . Thus, the solution shows the model Eq. (1) with AFM  $J_3$  and FM  $J_1$  has

captured the main features for an incommensurate correlation in  $\text{NiGa}_2\text{S}_4$ , but it is still oversimplified as the minimal model for all low temperature properties of  $\text{NiGa}_2\text{S}_4$ . A biquadratic interaction might be a good candidate for reproducing a finite susceptibility at zero temperature. In the absence of the 3rd-NN interactions, a biquadratic term can induce a quadrupolar order and totally suppress the spin order. The  $T^2$ -law of specific heat is also intact when quadrupolar order sets in [19, 20, 21]. It will be interesting to see how the incommensurate spin correlation be influenced by the biquadratic interactions.

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